Los Alamos
NATIONAL LABORATORY
EST. 1943

LA-UR-19-24916
Approved for public release; distribution is unlimited.

Title: Initial Assessment for a Generic Horizontal Fissile Solution Vessel with an Electron Beam-based Neutron Generator

Author(s): Kim, Seung Jun
Verner, Kelley Marie

Intended for: Report

Issued: 2019-05-29

Disclaimer:
Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by Triad National Security, LLC for the National Nuclear Security Administration of U.S. Department of Energy under contract 89233218CNA000001. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Initial Assessment for a Generic Horizontal Fissile Solution Vessel with an Electron Beam-based Neutron Generator

Seung Jun Kim, Kelley Verner
Nuclear Engineering and Nonproliferation Division
Nuclear System Design and Analysis (NEN-5)
Los Alamos National Laboratory
Los Alamos, NM 87544

Executive Summary

The LANL NA-23 program office has provided an extensive design study for a medical isotope (i.e. Molybdenum-99) production facility based on a fissile solution vessel with various accelerator technologies. These concept designs and associated R&D have provided affordable pathways for system optimization for industry counter partner such as SHINE and Global Medial Isotope Supply (GMIS). A horizontal fissile solution vessel design with an electron beam (E-beam) accelerator is proposed and studied with modeling capabilities developed for previous concepts (i.e. a generic vertical fissile solution vessel with a deuterium-tritium (DT) accelerator). An initial system assessment is performed by estimating theoretical Molybdenum-99 production yield with MCNP6.2. A baseline system configuration is constructed based on early design discussions between LANL and GMIS. Since only an initial design concept is available at this stage, all associated system design parameters can be adjusted to fit the best system performance characteristics. In this work, unlike previous vertical system studies, the cooling configuration of the horizontal fissile solution vessel is not considered, and only fission reactions are used to quantify the theoretical product yield. Five parametric design studies are proposed, preliminary results are briefly reported, and extended design studies beyond what is proposed here are also discussed. It is concluded that a comprehensive system analysis via multiphysics coupled calculation developed by LANL [3,4] should be further conducted for the final design.

Contents

Figures

Tables

1. Background on Mo99 production methods using nuclear technology

Molybdenum-99 (Mo-99) is a critically important radioactive material used in medicine. The decay product of moly-99, Technetium-99m (Tc-99m), is used worldwide for medical diagnostic imaging of many types of cancer, heart conditions, and other diseases. The need for domestic Mo-99 production is discussed thoroughly in the literature [1-5]. This motivation can be briefly summarized as follows: (1) the US consumes about half of the world's supply of Mo-99, but has had no domestic production since the late 1980s; (2) therefore, the US imports Mo-99 from Canada, Australia, Europe, and South Africa; and (3) this condition situates the US in a medical crisis that directly relates to national security.

Traditionally there are three production methodologies used to create Mo-99. First is LEU fission-based production: U235(n,f). In this method, a neutron interacts with fissile nuclei, largely LEU material, to start a fission reaction. Mo-99 is generated as a byproduct of this reaction. The second method is neutron capture: (n,γ). A neutron is captured by Mo-98 nuclei and becomes Mo-99. The third method is accelerator-based production: (γ,n). High velocity electrons interact with Mo-100 (or other heavy Z atom) to generate bremsstrahlung gamma rays. High energy gamma rays interact with Mo-100 to produce a neutron and Mo-99 as byproducts. The production yield of these three methods are highly dependent of target nuclei enrichment (i.e. uranium enrichment in method 1 or Mo-98 and Mo-100 enrichments in method 2 and 3) as well as separation efficiency. In addition, Mo-99 production methods can be categorized by system design approach. One is a fission reactor based (n,f) or (n,γ) approach, and the other is an accelerator based approach. Figure 1 illustrates the aforementioned production methodologies. Recently, an alternative Mo-99 production method that utilizes complementing advantages from each technique has been developed. An example of this is a fissile solution reactor combined with novel neutron generator concepts (i.e. deuterium – tritium (DT) or E-beam accelerators). This study will focus on alternative Mo-99 production methods by evaluating the practicability of the proposed system design and quantifying technical challenges.

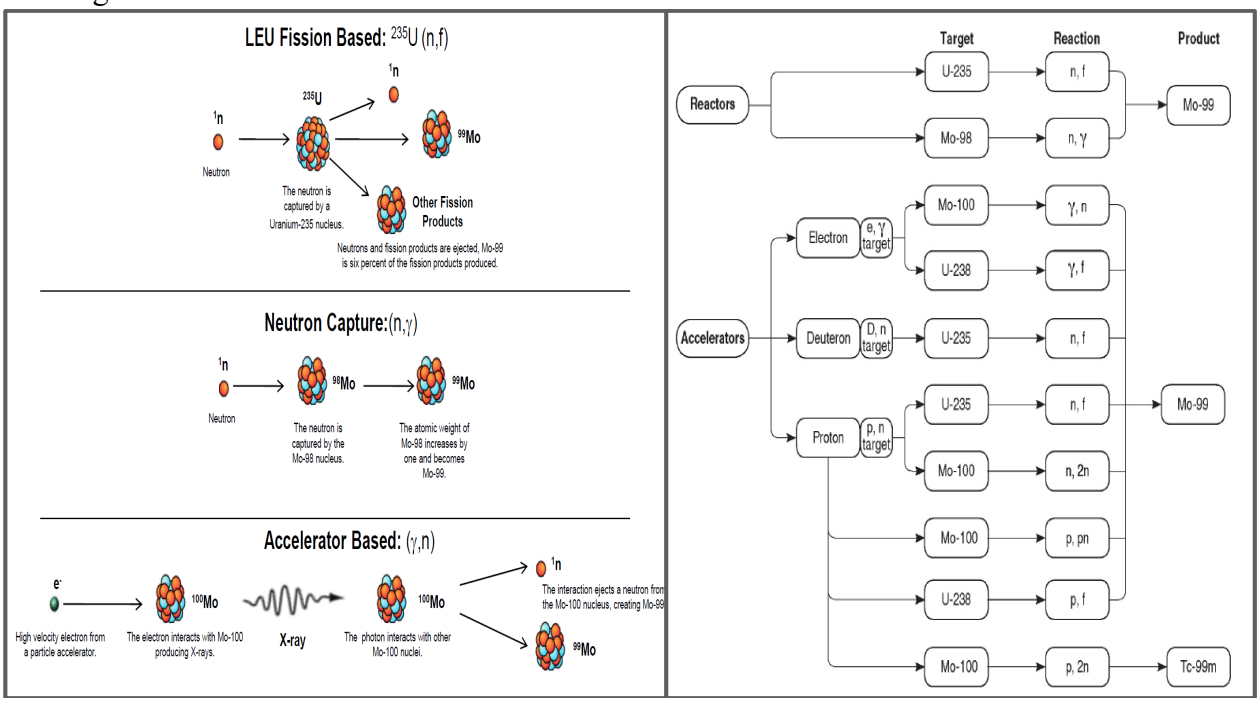

Figure 1 Mo-99 production methodologies using reactor and accelerator technique [2]

2. System Configuration of Horizontal Solution Vessel

2.1 Overall design descriptions

Based on LANL's extensive expertise on generic solution vessel designs for Mo-99 production, a new concept design is proposed to conduct an initial system assessment for the company, Global Medial Isotope Supply (GMIS). In this concept design, a generic horizontal solution vessel is designed in conjunction with a horizontal E-beam orientation instead of vertical solution configuration previously used for the SHINE. Mo-99 production via E-beam based fission was initially proposed by Argonne National Laboratory (ANL). A prototypic, scale production facility, which included a target isotope separation capability, was demonstrated in ANL's team [6].

A detailed system configuration of the proposed generic horizontal solution vessel is illustrated in

Figure 2. The fissile solution resides in the horizontal vessel that includes a degassing region for fission gas or any form of radiolytic by-production in the tank. Highlighted in a blue color in both front and side view in

Figure 2 is the fissile solution, while the degassing region is in green. The E-beam accelerator is featured with a 40 Mev, 100kW, horizontal beam. For neutron generating purposes, a natural uranium target is positioned along with the E-beam line. It is designated with the yellow color. The shape and location of the target can be adjusted to maximize the photonuclear reaction rate. Two options of acid bases for the fissile solutions were used; uranyl nitrate and uranyl sulfate. In the degassing region, radiolytic gas as well as potential fission gas can be stripped off by using inert gas (e.g. argon).

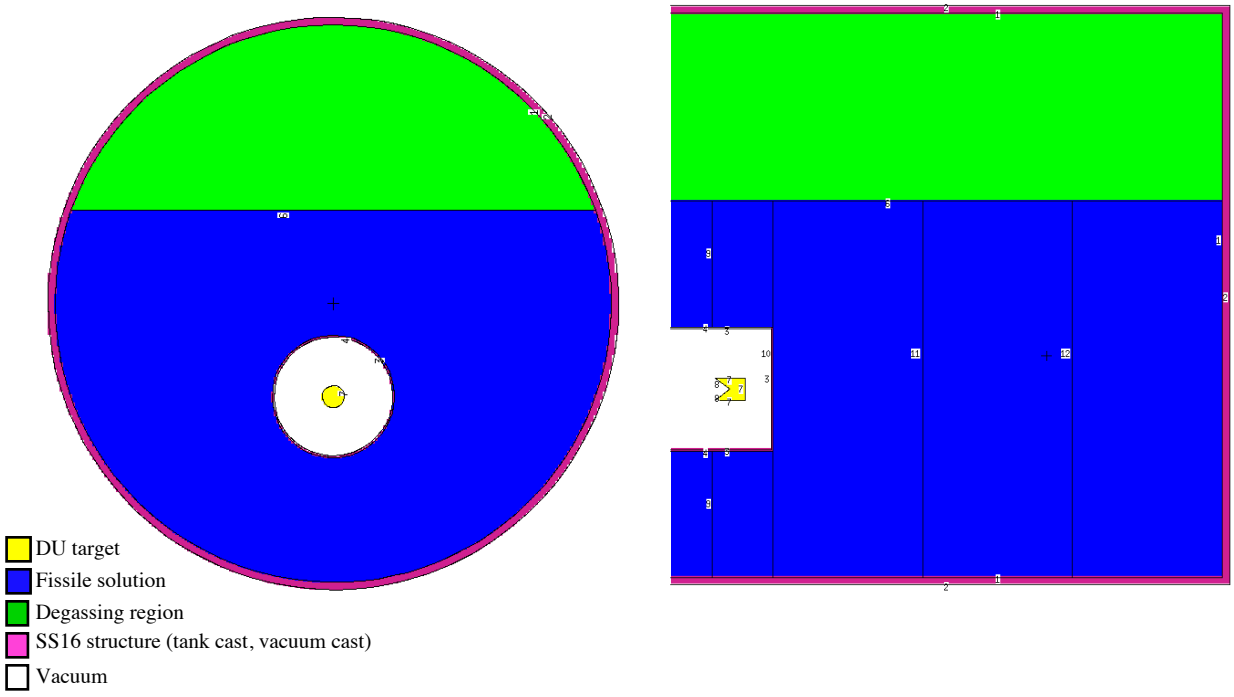

Figure 2 Schematic diagrams of generic solution design (Left: front view, Right: side view)

In Figure 2 above the corresponding colors are associated with different aspects of the system:

- Yellow: DU target
- Blue: fissile solution
- White: vacuum
- Pink: SS316 structural tank
- Green: degassing region

Convective heat transport (i.e. thermal mixing) by fission induced heating and heat removal by cooling channels is not considered at the current stage of conceptual design. However, those cooling configurations should be implemented for a full system analysis once associated thermal hydraulic characteristics are decided.

2.2 Tank specification

The fissile solution contained the horizontal cylinder vessel has a target volume of approximately 1000 L not including the extra space for the degassing area. The detailed geometry information used in the current study is summarized below.

- Tank diameter: 150 cm
- Tank shielding thickness: 2 cm
- Tank horizontal length: 150 cm
- Vertical solution height: 100 cm
- Solution Volume: 1012 liter

2.3 Material

Materials used in the current solution vessel are listed below. For simplicity, only four key materials are modeled in the initial MCNP assessment. Relevant density information is also reported.

- Shielding material for vessel: SS316, 7.9 g/cm^3
- Solution: Uranyl Nitrate, 1.3 g/cm^3
- Target: Natural Uranium, 18.95 g/cm^3
- Upper void: Air, 5E-4 g/cm^3

2.4 Vacuum chamber specification

A cylindrical vacuum chamber is located in the left region of solution tank and guides the electron beam toward to the target material for neutron generation. The location and size of the vacuum chamber can be adjusted to maximize the theoretical Mo-99 production yield while not minimizing the fissile solution volume. Detailed cooling mechanisms on the vacuum chamber as well as appropriate thermal management for the target are out of the current scope of the work. Relevant geometry information regarding the vacuum chamber is listed below.

- Vacuum chamber length: 30 cm
- Vacuum chamber diameter: 32 cm
- Chamber shielding thickness: 0.5 cm

2.5 Target specification

A unique aspect of this design is that photonuclear (γ, f) reactions are used to produced neutrons

with a natural uranium or depleted uranium target. As the photonuclear cross section is notably smaller compared to other reaction cross sections, efficient target design for neutron generation is one of key components determining the overall production yield. Below are the baseline designs utilized in the initial system assessment. There is room for improvement on the target design and will be explored in later work.

- Target diameter: 6 cm
- Target length: 8 cm
- Corn depth: 4 cm
- Target depth: 15 cm

In order to improve the target design, such parameters as shape, location, material selection, and multi-disk array for cooling strategy can be further investigated for higher fission rates in the proposed horizontal fissile solution vessel. A schematic of the target configuration in the vacuum chamber and potential parametric study proposed for the next study is illustrated in Figure 3.

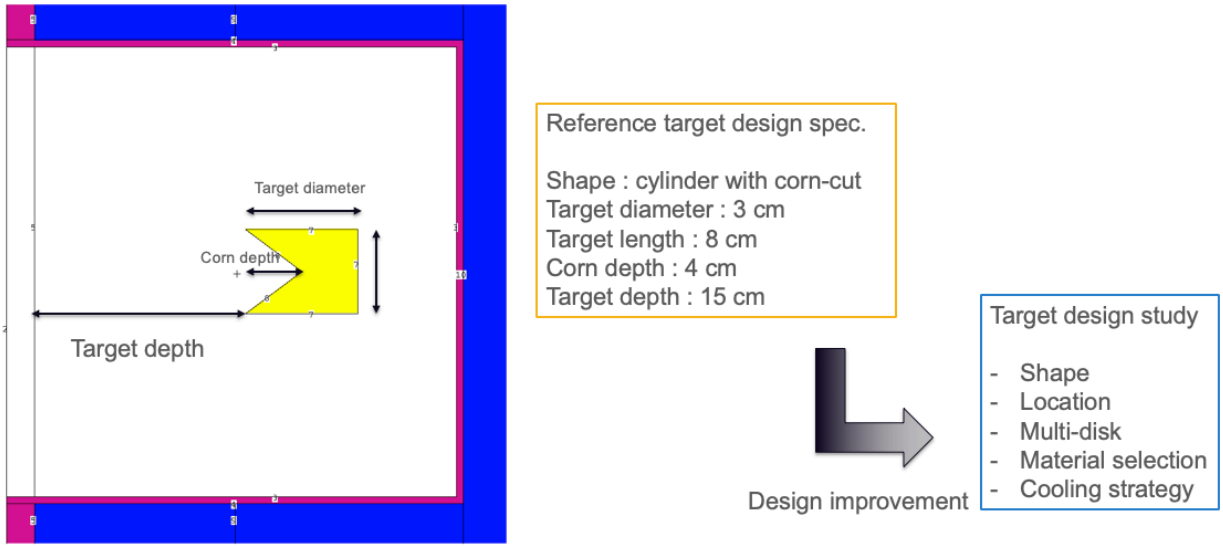

Figure 3 A close-look on a target in the vacuum chamber and possible target design study

3. Design Requirement for Multiphysics Coupled Calculation

One of unique characteristics of a fissile solution system is an inherent physics coupling between neutronics and thermal hydraulics. In the operating process, thermal expansion from the fission induced heating with radiolitic gas (void) generation will impact the solution density as well as temperature profile which provides effectively negative feedback on the fission reaction that makes the system behave in a favorable manner in both normal and abnormal conditions. Besides this inherent coupling phenomena, a fission reaction in the solution acts as a heat source while an engineered cooling configuration within the solution tank acts as heat-sink to create a natural convection heat transfer system. The Multiphysics coupled calculation is developed and fully demonstrated in a generic vertical solution vessel study [see reference 3,4, and 5]. In the current study, the cooling configuration is not quantified at the initial design level. Therefore, there is no coupled calculation performed to evaluate the system behavior at any given operating condition.

Instead, we are focusing on the production yield based on the proposed design with horizonal orientation vessel using MCNP calculation.

Once the initial assessment is complete, a series of cooling configurations can be investigated based on the industry partner's target performance level. However, the author recommends that additional thermal hydraulic considerations and constraints be accounted for when the cooling configuration is designed. The following items are the focus areas for future studies if resources are available.

- Cooling tube location and orientation
- Natural convection or forced convection strategy in the vessel
- Bubble induced turbulence effect on the convective heat transfer coefficient
- Overall HTC calculation
- Degassing related thermal hydraulic consideration

4. Methodology

4.1 MCNP

Using the baseline design geometry described in section 2, the neutron transport simulation is performed under variety of solution conditions to evaluate the fission rate in the fissile solution. MCNP6.2, with the ENDF/B-VII library, is used in this calculation. MCNP is a general-purpose Monte Carlo N-Particle code used for various particle transport applications. MCNP provides both the free gas and S(α, β) model for the thermal neutron cross section library. Since the fissile material is dissolved in the aqueous solution (i.e. uranyl nitrate ($UO_2(NO_3)_2$ or uranyl sulfate (UO_2SO_4), accounting of appropriate thermal neutron effect on the fission reaction is believed to be critical for high fidelity calculation. Here, we use the S(α, β) model-based library to adequately apply the thermal scattering effect on the light elements, such as hydrogen, in the aqueous solution.

4.2 E beam characteristics

Two different electron beam characteristics are considered here. One is 40Mev with 100kW power which leads to the corresponding beam intensity of 1.56E16 [e/s]. The other is 35Mev with 10kW power that is equivalent to the beam intensity of 1.78E15 [e/s]. These two cases are selected to meet GMIS's design requirements and the existing beam characteristics of ANL. However, the feasibility of those accelerator's beam characteristics is beyond the current scope of assessment.

4.3 Uranyl solution density data

The uranyl nitrate solution density was measured as a function of molar concentration and documented in The Oak Ridge National Laboratory report [7]. The experimental data available in the report covers up to 1 mole/liter of uranyl nitrate. Past this value, the corresponding solution density is extrapolated linearly based on the existing experimental data. The estimated density is only extrapolated before the solution's solubility limit assuming that precipitation (i.e. unexpected particulate forming in the solution) is not taking place at this density measurement. The concentration versus density of uranyl nitrate is plotted in Figure 4. The corresponding density information for uranyl sulfate for case 6 is extracted from the FY17 Milestone report [4].

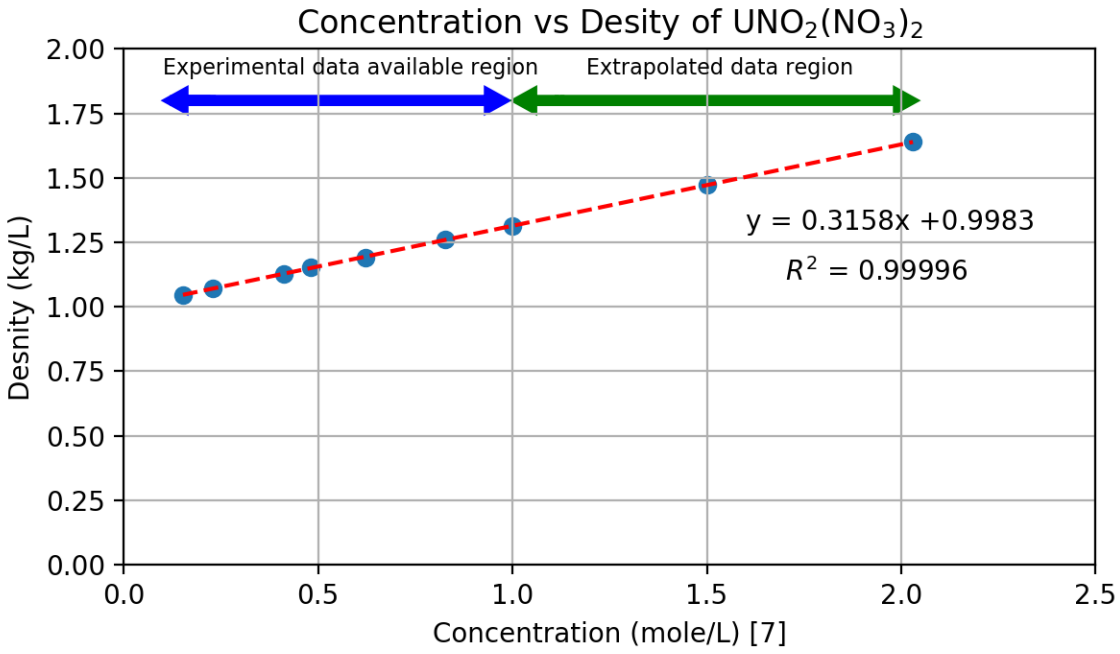

Figure 4 Measured and correlated uranyl nitrate solution density against molar concentration

Detailed calculation notes for the molar concentration of uranyl nitrate or sulfate can be found in appendix A and the previous milestone report [3]. It should be noted that the solution density in the fissile solution analysis is a most critical parameter in determining the criticality and fission rate. Because the calculated solution density was extrapolated from 1950s experimental data, an updated density measurement with different concentration levels of uranyl nitrate solution would be highly desirable for high fidelity production yield calculation.

4.4 Mo-99 production yield calculations

The Mo-99 production yield at the target irradiation time is estimated using the activation equation along with the MCNP-informed fission rate and the theoretical production yield for Mo-99.

$$Isotope\ production\ yield\ [t] = A_o\left(1 - exp^{-\lambda t}\right) \qquad \text{eq. 1}$$

$A_o, \lambda,$ and t denote the initial activity of target isotope, the half-life, and the target irradiation time. In this assessment, the target irradiation time is set to be 100 hours, excluding any loss from the separation process after the irradiation. The initial activity of the Mo-99 [#/s] is calculated from the fission rate [#/source particle], source particle intensity [SP/s] from E-beam accelerator, and theoretical production yield of the target isotope (approximately 6.01%).

4.5 Keff values and subcriticality

Note that the key design goal of combining a generic fissile solution with an accelerator is to control the irradiation easily to prevent achieving criticality as well as controlling the criticality in the solution. This means the system must always be subcritical in any operating condition. As shown in **Error! Reference source not found.**, the Keff values are all below unity, which makes the system subcritical. However, from the production yield perspective, the lower the criticality, the less production yield achieved. Therefore, the correlation between Keff and the target production

yield must be optimized based on the overall system economics and performance. In addition, as the Keff is a safety margin related parameter, any associated safety considerations (i.e. solution boiling by overheating at high Keff) should also be further investigated. In this study, the subcriticality for each test condition are confirmed only. The optimal Keff value for production yield is not sought in the assessment process.

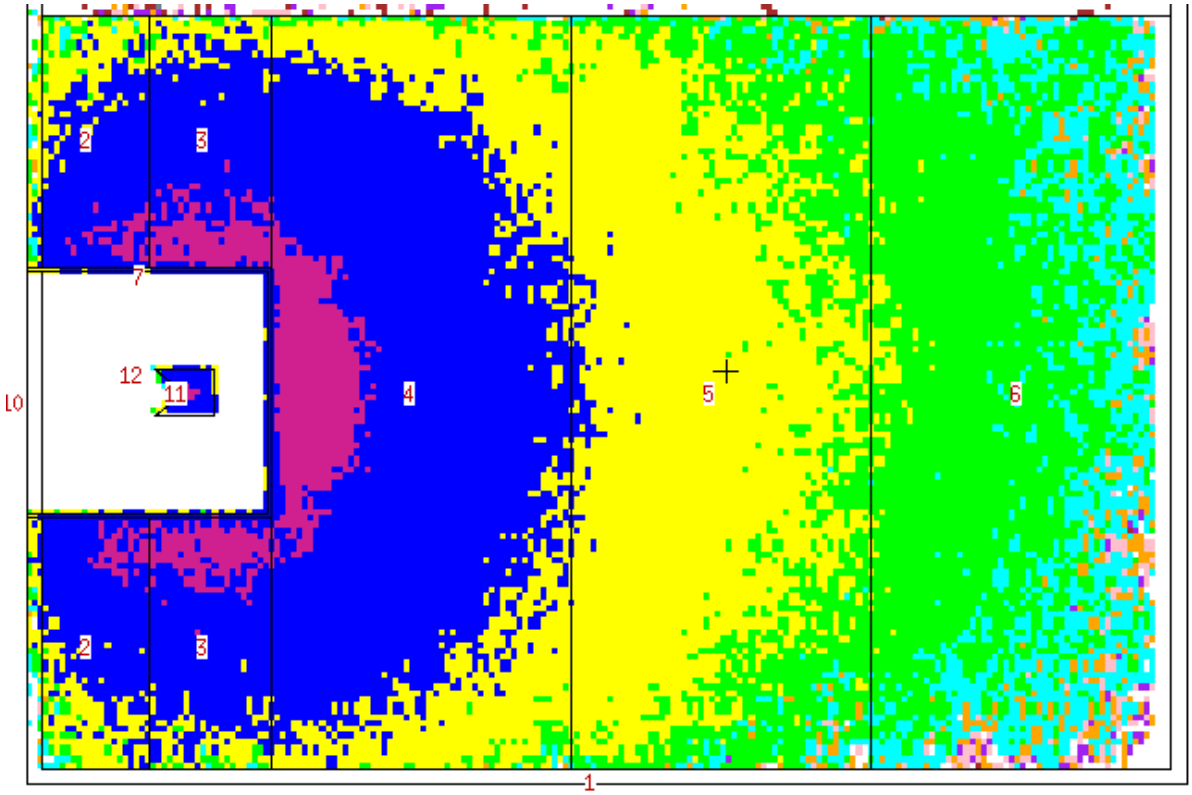

Figure 5 A fission rate contour in the solution using F4 tally

4.6 Fission rates and MCNP tallies

Fission rates are estimated by employing F4(-1,2,-6) , and F8(-1,2,-1,-4) tally in the MCNP calculation. The F4 tally provides a total fission rate in the cell which can be directly converted into the Mo-99 production rate. An F8 tally can evaluate the Mo-99 residual in the cell by accounting all of associated decay reaction toward to Mo-99 from neighboring isotope. The F4 tally-based Mo-99 production rates and F8 tally-based Mo-99 residuals are compared and are in reasonable agreement. The total production rate by the F8 tally method provides approximately 8% higher Mo-99 production rate compared to the F4 tally. The increase can be attributed to the neighboring isotope decaying effect which is not accounted for in the F4 tally that only counts the fission rate.

4.7 Sectioning tallies of fissile solution vessel

In addition to the total production yield, bisecting the solution vessel into 5 different regions produces a sectional Mo-99 production yield within the solution vessel. While Figure 5 illustrates only qualitative fission rate across the entire solution vessel, Table 1 and Figure 6 can provide a

quantitative fission rate in each sectional zone in the solution. As expected, most of the Mo-99 production is observed at section 2 and 3 with 32.88% and 45.26%, respectively. Please note that fluid mixing in the solution vessel is not accounted for in this MCNP calculation. It is interesting to point out that the departed region from the E-beam chamber, such as section 6, is producing a Mo-99 yield less than 1% even though the volume of section 6 is the largest. This finding should be further investigated when the thermal hydraulic and neutronic coupled calculations are performed in the next design study.

Table 1 Sectional production yield using F4 and F8 tally and corresponding heating calculation

F-Tally	Tally description	Region-2	Region-3	Region-4	Region-5	Region-6	total
F24(-1,2,-6)	total fissionrate in cell-# [#/SP]	5.36E-03	1.04E-02	1.43E-02	1.41E-03	1.30E-04	3.16E-02
	percentage [%]	16.96%	32.88%	45.28%	4.46%	0.41%	100.00%
				Mo99=	Totall fission*0.6%=		1.93E-03
F24(-1,2,-1,-4)	total fission induced heating in cell# [Mev/SP]	9.37E-01	1.81E+00	2.50E+00	2.46E-01	2.27E-02	5.52E+00
	percentage [%]	16.99%	32.88%	45.26%	4.46%	0.41%	100.00%
FT8	Mo99 residual in cell-# [#/SP]	3.54E-04	6.93E-04	9.48E-04	9.67E-05	8.98E-06	2.10E-03
	percentage [%]	16.85%	32.98%	45.14%	4.60%	0.43%	100.00%
					M99= Total residual Mo99 rate=		2.10E-03

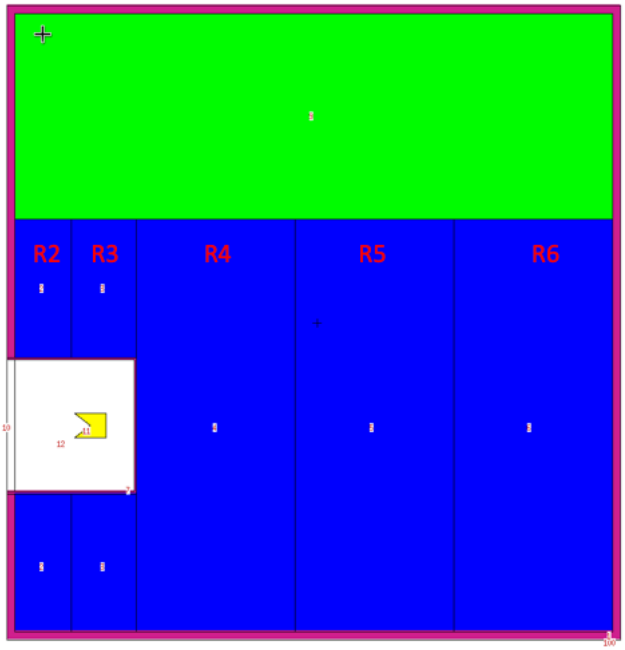

Figure 6 A schematic diagram with regional naming convention (R2, R3, R4, R5, and R6)

4.8 Simulation design and test matrix

Six different solution conditions are chosen to evaluate the criticality and fission rate in the horizontal solution vessel. In the test matrix (See Table 2), uranyl solution type, enrichment level, solution concentration, and solution base are varied as parametric variables. The solution density is estimated from the experimental correlation of solution thermo-physical properties. First, a

KCODE calculation is conducted for each test condition to see what subcriticality is achieved for the corresponding solution scenarios. Then, an MCNP calculation with a reasonable number of source particles [NSP = 1million] is performed with the NEN-5 group cluster using an adequate message passing interface (MPI) setting. Both KCODE and MCNP calculations require a viable target solution density as a key input. Table 2 summarizes the Keff and Mo production yield results based on 40Mev and 100kW beam conditions.

The proposed test matrix carefully quantifies the following aspects on the system design.

- Solution base selection effect on the production yield
- Solution concentration effect
- Solution enrichment
- Solution type selection (Uranyl Nitrate vs Uranyl Sulfate)
- Accelerator beam characteristics effect

Table 2 LANL proposed parametric test condition for various solution scenarios

	Uranyl Type	*Uranium Enrichment*	*Solution Concentration*	*Solution Base*	*Sol. Density[g/cc]*	*Keff*	*Mo99@100hrs [Curies]*
Case 1	$UO_2(NO_3)_2$	0.7% (Nat.)	1.015 M/L (400g/Liter)	H_2O	1.31	0.1981	30
Case 2	$UO_2(NO_3)_2$	0.7% (Nat.)	1.015 M/L (400g/Liter)	D_2O	1.31	0.4163	85
Case 3	$UO_2(NO_3)_2$	0.7% (Nat.)	1.523 M/L (600g/Liter)	H_2O	1.47	0.2607	46
Case 4	$UO_2(NO_3)_2$	0.7% (Nat.)	2.031 M/L (800g/Liter)	H_2O	1.64	0.3217	60
Case 5	$UO_2(NO_3)_2$	3% (Nat.)	2.031 M/L (800g/Liter)	H_2O	1.64	0.8591	724
Case 6	$UO_2(SO_4)$	0.7% (Nat.)	0.588 M/L (215g/Liter)	H_2O	1.185	0.12	18

5. Preliminary results of initial system assessment

5.1 H_2O vs D_2O effect

Figure 7 provides a Mo-99 activity estimation over 100 hrs of irradiation with the conditions from Table 1 at accelerator specification of 40 MeV and 100kW. Among the six different cases studied, the results from 5 cases are plotted as a function of irradiation time. When Case 1 and Case 2 are compared, the effects of the solution base are clearly seen. The two cases have the same solution conditions but not the same base solutions. One uses light water and the other uses heavy water. It is apparent that the heavy water solution base produces notably higher Mo-99 activity compared to the light water-based solution. From a cross-section standpoint, heavy water has a lower absorption cross section which leads to better thermalizing in the solution. It is evident that heavy water serves as a better moderator with a higher fission rate. However, other operating constraints have to be considered when heavy water is used. One of important safety and licensing concern associated with heavy water is the non-trivial amount of tritium produced from the deuterium-neutron reaction. This undesirable isotope production requires an additional off-gas system design to be included in the fissile solution vessel system and adds more layers of licensing challenges, hampering the system operating efficiency. Therefore, in the current study, the D_2O based solution (Case2), is intentionally excluded in the assessment even though it can provide a higher fission rate in the solution vessel.

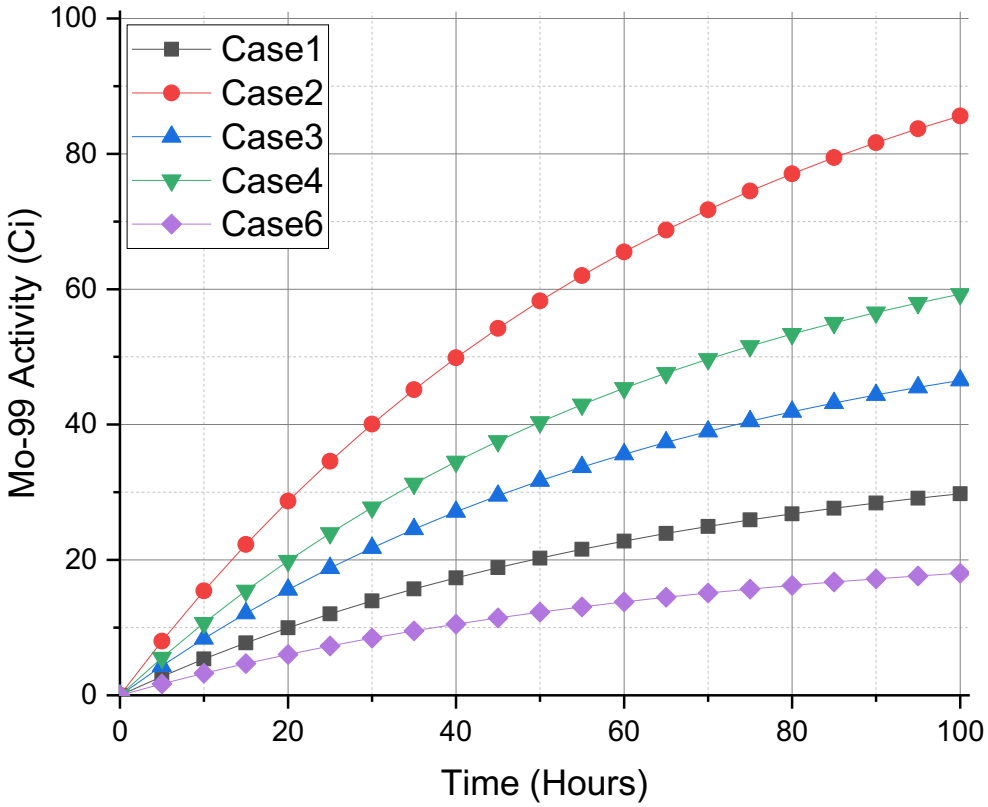

Figure 7 Mo-99 activity calculation over the 100 hours of irradiation time with 5 different solution scenarios (Case 5 is excluded in this figure as the Mo-99 activity falls into a different order of magnitude)

5.2 Uranyl nitrite concentration effect

With Case 1, 3, and 4, the concentrations of uranyl nitrite are 400g/L, 600g/L, and 800g/L, making the molar concentrations, 1.015 mole/L, 1.523 mole/L, and 2.03 mole/L. As expected, the Mo-99 production yield is linearly proportional to the solution concentration. Which leads to the following questions: 1) What is the maximum achievable uranyl nitrite concentration and 2) What is the solubility limit of the uranyl nitrite in the target solution? Solubility of uranyl nitrate or uranyl sulfate are key properties needed to perform a wide range of parametric studies. For the purposes of this work, the maximum concentration of nitrate solution is bound to less than 2.1 mole/L.

5.3 Enrichment effects

Natural uranium contains about 0.7% U235 and is used in the Case 4 calculations. Whereas, Case 5 uses uranium at 3% enrichment. To illustrate how U235 enrichment affects the Mo-99 yield, these two cases were compared with differing enrichments while leaving all other variables identical. KCODE calculations indicate that the 3% enriched solution (Case5) has a Keff of 0.8591 (i.e. still subcritical), while the Case 4 produces a Keff of 0.3217. Interestingly, the Mo-99 production yield at 100 hours with the 3% enrichment case is estimated to produce 724 Curies of Mo-99 activity. These results indicate 3% enriched case generates 12 times more yield than the natural uranium

case even though all other parameters are held constant. It is clear that enriched uranium in the solution is a promising strategy to meet the industry's performance goal.

5.4 Uranyl nitrate vs uranyl sulfate effect

The effect of solution type on the production yield is also evaluated by comparing Case 1 and Case 6. When the molar concentration of the applied solution is similar, the production yields for both nitrate solution and sulfate solution are comparable. No noticeable change is observed in the current study. However, as the two solutions exhibit noticeable differences in the solubility limit (usually, uranyl nitrate has higher solubility than uranyl sulfate), the nitrate-based solution would be more preferable for the higher yield performance. Solubility and pH control are crucial chemical properties which determine the feasible operating window for a fissile solution application. Related literature is reviewed and summarized in the FY18 milestone report [8].

5.5 Accelerator power effect

Next, the same six cases of solution scenarios are applied with a reduced electron beam accelerator power (35 Mev/ 10 kW). When comparing the results from Table 2 and Table 3, it is evident that the beam power is proportional to the yield.

Table 3 LANL proposed parametric test condition with reduced beam power

	Uranyl Type	*Uranium Enrichment*	*Solution Concentration*	*Solution Base*	*Sol. Density[g/cc]*	*Keff*	*Mo99@100hrs [Curies]*
Case 11	$UO_2(NO_3)_2$	0.7% (Nat.)	1.015 M/L (400g/Liter)	H_2O	1.31	0.1981	**3.4**
Case 12	$UO_2(NO_3)_2$	0.7% (Nat.)	1.015 M/L (400g/Liter)	D_2O	1.31	0.4163	**9.7**
Case 13	$UO_2(NO_3)_2$	0.7% (Nat.)	1.523 M/L (600g/Liter)	H_2O	1.47	0.2607	**5.2**
Case 14	$UO_2(NO_3)_2$	0.7% (Nat.)	2.031 M/L (800g/Liter)	H_2O	1.64	0.3217	**6.8**
Case 15	$UO_2(NO_3)_2$	3% (Nat.)	2.031 M/L (800g/Liter)	H_2O	1.64	0.8591	**82.5**
Case 16	$UO_2(SO_4)$	0.7% (Nat.)	0.588 M/L (215g/Liter)	H_2O	1.185	0.12	**2.1**

5. Summary and suggested future work

This report aims to understand the design parameters of a generic horizonal vessel with an E-beam accelerator used for Mo-99 production. A baseline system design is defined, and detailed dimension information is addressed for an initial system assessment. The objective function of the current parametric study is solely to maximize the theoretical Mo-99 production yield at 100 hours of accelerator operation time. No other operational safety parameters (i.e. fissile solution temperature, radiolytic gas transport, or thermal hydraulic related considerations) are assessed at this point. Preliminary results from the initial assessment of the baseline system are summarized below, and possible suggestions for the extended study are described as follows.

The summary and lessons learned:

- A generic horizontal solution vessel with E-beam accelerator for a Mo-99 production facility is proposed. A baseline vessel design and associated E-beam characteristics are defined for a wide range of parametric studies for differing fissile solution scenarios.

- An MCNP model of the system configuration and solution conditions is created to evaluate the criticality and fission rate in the solution.
- Uranyl nitrate is used as a starting selection for the fission solution material. A horizontally oriented E-beam accelerator is designed to produce neutrons by either (γ,n), (γ,f) reactions in the target material (i.e. natural uranium or depleted uranium)
- The cooling configuration is not determined at the current design stage, no Multiphysics coupled calculation (MCNP+CFD) are performed.
- The use of a heavy water solution provides a higher fission rate due to its lower absorption cross section. This leads to more efficient thermalization. However, other safety concerns could arise due to undesirable tritium generation in the heavy water solution.
- Solution concentration appears linearly proportional to the fission rate (i.e. Mo-99 production yield). Note that the solution concentration can be increased only up to its solubility limit at the operating temperature and pH conditions. Therefore, solution chemistry needs to be carefully monitored. The literature reports that the solubility of uranyl nitrate is relatively higher than that of uranyl sulfate. This implies that uranyl nitrate based-fissile system might be operated with relatively higher solution concentrations.
- 3% enriched Uranium solution (Case5) generates 12 time more Mo-99 production yield compared to the natural uranium solution (Case 4).
- No significant production yield change is observed between uranyl nitrate and uranyl sulfate
- Two different accelerator conditions are tested, and the electron beam intensity is linearly proportional to the production yield in all of solution scenarios. A feasibility study of the tested accelerator condition over the target irradiation time (i.e. 100 hrs) is beyond of the current scope of study.

An extended parametric test matrix is proposed below to investigate further studies.

- Target location study: 15, 45, 75, 105, 125 cm away from the left side of tank wall
- Target shape study: corn-shaped cut, multi-disk concept (best design in term of target cooling performance)
- Solution volume change by varying the size of the vacuum chamber
- Potential cooling configuration idea development
- Solution mixing phenomenon study with a proposed cooling configuration design

6. References

1. Paula Gould, “Medical isotope shortage reaches crisis level” Nature 460, 312-313 (2009).
2. IAEA Nuclear Energy series, “Non-HEU production technologies for molybdenum-99 and Technetium-99m” IAEA No. NF-T-5.4, 2013.
3. S. Jun Kim, Cynthia Buechler, “A design study of generic solution vessel systems”, LA-UR-18-05-09
4. S. Jun Kim, Cynthia Buechler, “Demonstration of multi-physics (neutron transport and thermal hydraulic) coupling methodology for liquid-fuel based nuclear technology application” Transaction of the ANS winter conference, 2018, vol. 117, pp 1764-1797
5. S. Jun Kim, Cynthia Buechler, “A fully coupled MCNP and multiphase CFD for a fluid fuel subcritical system” Transactions of the ANS conference, 2017, LA-UR-17-29641

6. A. J. Youker, et al., “A Solution-Based Approach for Mo-99 Production: Considerations for Nitrate vs. Sulfate Media” Science and Technology of Nuclear Installations, 2013, ID:402570
7. J.L. Botts, et al., “Density, Acidity and Conductivity Measurements of Uranyl Nitric Acid Solutions” ORNL/TM-6491, 1952.
8. Mario R. Naranjo, R. Blake Wilerson, Seung Jun Kim. “The Argus Solution Reactor and Molybdenum Production: A Summary Report Based on Open Literature”, LA-UR-18-12923

Appendix A: *A calculation note for atomic fraction of uranyl nitrate constituents at a given solution concentration and density*

Assume that solution concentration [gU/L] and the associated fissile solution density [g/L] are known values. One can calculate the atomic fractions of the constituents of the uranyl sulfate solution, which are key input parameters for the MCNP calculation. The chemical formula of the uranyl sulfate solution is $UO_2(NO_3)_2 + H_2O$. At the given solution concentration [g/L], the mole per liter of solution atoms can be calculated as shown below while assuming the uranium is NOT enriched (i.e. Natural Uranium). Assume that solution concentration is 400 g/L in this calculation note.

Mole per liter of $UO_2(NO_3)_2$ [mole/L] = $UO_2(NO_3)_2$ [g/L] / molar mass [g/mole]

400 g/L / 393.979 g/mole = 1.015 mole/L

With the chemical formula, mole per liter for each constituent of the $UO_2(NO_3)_2$ can be calculated as follows.

U = 1.015 * 1 mole/L

O = 1.015 * 8 mole/L

N = 1.015 * 2 mole/L

With calculated values for mole per liter and known molar masses, the mass per liter for each constituent is consecutively obtained.

Mass of U per liter = 237.977 g/mole * 1.015 mole/L = 241.616 g/L

Mass of O per liter = 16 g/mole * 1.015 *6 mole/L = 129.956 g/L

Mass of N per liter = 14 g/mole * 1.015 * 2 mole/L = 28.428 g/L

The total mass of $UO_2(NO_3)_2$ (i.e. solute) per liter is 400 g/L. The total mass of H2O (i.e. solvent) per liter can be calculated provided the solution density is given.

Using multivariable regression method, a polynomial correlation as a function of concentration and temperature is developed and documented in Figure 4. At 1.015 mole/liter, the solution density is assumed to be 1310g/L. This density value allows the total mass of H2O per liter to be calculated as 910 g/L. Thus, the mole/L for each constituent of H2O can be written

Mole/L for H2O = 910 [g/L] / 18 [g/mole] = 50.55 mole /liter

Thus, the mole of hydrogen and oxygen per liter are determined to be 101.11 and 50.55 mole/liter. The mole per liter can be converted into the mass of each constituent of H2O.

Mass of H per liter = 1 g/mole * 101.11 mole/L = 101.11 g/L

Mass of O per liter = 16 g/mole * 50.55 mole/L = 808.89 g/L

Mass of H2O per liter = 101.11+808.89 = 910 g/L

Now we obtain every individual constituent mole value in a unit volume, which provides us a molar fraction of the constituent as listed below.

Uranium = 1.015 mole/liter

Hydrogen = 101.11 mole/liter

Oxygen = 55.55 + 6.09 = 61.64 mole/liter

Nitrogen = 2.03 mole/liter

The fractional ratio of isotopic element is determined based on the nuclear wallet cards database version of 3/31/2018. Refer to the link here https://www.nndc.bnl.gov/nudat2/indx_sigma.jsp

After the fractional isotopic element ratio is considered, the molar fraction (i.e. atomic fraction) for each constituent is listed above. These calculated molar fractions are to be an input for the MCNP material card for the associated solution concentration. Note that the molar fraction values are automatically normalized in MCNP even if the atomic ratios are entered in a non-normalized fashion.

Appendix B: *MCNP input deck for Case 5, criticality calculations*

AQUEOUS HORIZONTAL SUB-CRITICAL EXPERIMENT

```
c CELL CARDS-------------------------------------------------------------------
1 1 -7.9  1 -2 4 imp:n,e,p=1 $ LARGE CYLINDER CELL
2 2 -1.31 -1 -9 -6 4 imp:n,e,p=4 $SOLUTION FILL CLOSEST TARGET SIDE (1)
3 2 -1.31 -1 9 -10 -6 4 imp:n,e,p=4 $SOLUTION FILL 2 CLOSEST TARGET SIDE (2)
4 2 -1.31 -1 10 -11 -6 imp:n,e,p=16 $TARGET PORT RIGHT SIDE (3)
5 2 -1.31 -1 11 -12 -6 imp:n,e,p=64 $SECOND FAR TARGET SOLUTION RIGHT (4)
6 2 -1.31 -1 12 -6 imp:n,e,p=256 $FARTHEST RIGHT SOLUTION AREA (5)
7 1 -7.9 -4  3 imp:n,e,p=1 $ TARGET CYLINDER CELL
8 0 -3  5 fill=100 imp:n,e,p=1 $ DEUTERIUM FILL CELL
9 4 -0.0005 -1  6 imp:n,e,p=1 $ AIR IN TANK
10 0 -3 -5 imp:n,e,p=1 $ cylinder face
11 3 -18.95 -7 8 u=100 imp:n,e,p=1 $ DU/NU TARGET
12 0 -8:7 u=100 imp:n,e,p=1 $ surrounding target void
100 0 2 -101 imp:n,e,p=1 $ VOID AROUND DECK
101 0 101 imp:n,e,p=0 $ END OF UNIVERSE

c SURFACE CARDS----------------------------------------------------------------
1 rcc 0 -8 0 0 150 0 75 $ inner cylinder macro
2 rcc 0 -10 0 0 154 0 77 $ outer cylinder macro
3 rcc 0 -10 -25.0 0 32 0 16 $ target source inner cylinder macro
4 rcc 0 -10 -25.0 0 32.5 0 16.5 $ target source outer cylinder macro
5 py -8 $ front fascia plane
6 pz 25 $ Solution fill plane
7 rcc 0 7 -25.0 0 8 0 3 $ target cylinder to be cut with cone
8 trc 0 7 -25.0 0 4 0 3 1e-05 $ cone cutout for cylindrical target
9 py 6.25 $ PLANE FOR CUTTING SOLUTION INTO PARTS
10 py 22.5 $ PLANE 2 FOR CUTTING SOLUTION INTO PARTS
11 py 62.333333333333336 $ PLANE 3 FOR CUTTING SOLUTION INTO PARTS
```

```
12 py 102.16666666666667 $ PLANE 4 FOR CUTTING SOLUTION INTO PARTS
100 c/y 0 -25.0 0.3 $ Constraint Tube for Gaussian (used for CCC cookie cutter cell)
101 rpp -500 500 -500 500 -500 500

c MATERIAL CARDS----------------------------------------------------------------
c mode n e p
c nps 1e4
c phys:p 100 J J 1 J J 0
c prdmp 1e3 1e3 0 20 1e3
c prdmp
c sdef dir=1 vec 0. 1. 0. x=d1 y=-33 z=d2 erg=40 par=e ccc=100
c sp1  -41  0.25479654  0 $ X gaussian spread, 0.6=fy=2.35482a
c sp2  -41  0.25479654 -25.0 $ Z gaussian spread, 0.6=fz=2.35482b
c f24:n 2 3 4 5 6 T
c fm24 (-1 2 -6)
c sd24 1 1 1 1 1 1
c fmesh4:n geom=xyz origin=-75 -10 -75
c          imesh=75 iints=160
c          jmesh=140 jints=200
c          kmesh=75 kints=160
c FM4 (-1 2 -6)
ksrc 0 13.0 -7.5
kcode 5000 1.0 50 250
c metal (boron 5010 and phos 15031 stripped for physics issue)
m1    14028.80c -0.001150761 16034.80c -2.33827E-06
      24053.80c -0.005492588 26054.80c -0.012563171 26058.80c -0.000645696
      28061.80c -0.000474334 42092.80c -0.001187171 42096.80c -0.001392605
      42100.80c -0.000837433 14029.80c -6.04609E-05 16032.80c -4.74334E-05
      24050.80c -0.002372672 24054.80c -0.001390266 26056.80c -0.204337425
      28058.80c -0.026936829 28062.80c -0.001535239 42094.80c -0.000756262
```

42097.80c -0.0008057 14030.80c -4.14207E-05

16033.80c -3.34038E-07 24052.80c -0.047530943 25055.80c -0.005010571

26057.80c -0.004805805 28060.80c -0.010731975 28064.80c -0.00040619

42095.80c -0.001315108 42098.80c -0.002056672

c heavy water uranyl solution with concetration of 80g/100ml (NU)

C m2 92235.80c 0.007 92238.80c 0.993 01002.80c 24.6237

c m2 92235.80c 0.05 92238.80c 0.95 01002.80c 54.70

C 07014.80c 2 08016.80c 20.3118

C mt2 hwtr.10t

m2 92235.80c 0.007 92238.80c 0.993

01001.80c 99.5891

08016.80c 57.7946

07014.80c 2

mt2 lwtr.10t

MX2:p j j 0 j j

c NU target (18.95 g/cm3)

m3 92235.80c -0.007 92238.80c -0.993

c air (wrong needs adjusting to proper air)

m4 8016.80c 1 7014.80c 2

Appendix C: MCNP input deck for Case 5, flux calculations

AQUEOUS HORIZONTAL SUB-CRITICAL EXPERIMENT

```
c CELL CARDS------------------------------------------------------------------
1 1 -7.9  1 -2 4 imp:n,e,p=1 $ LARGE CYLINDER CELL
2 2 -1.31 -1 -9 -6 4 imp:n,e,p=4 $SOLUTION FILL CLOSEST TARGET SIDE (1)
3 2 -1.31 -1 9 -10 -6 4 imp:n,e,p=4 $SOLUTION FILL 2 CLOSEST TARGET SIDE (2)
4 2 -1.31 -1 10 -11 -6 imp:n,e,p=16 $TARGET PORT RIGHT SIDE (3)
5 2 -1.31 -1 11 -12 -6 imp:n,e,p=64 $SECOND FAR TARGET SOLUTION RIGHT (4)
6 2 -1.31 -1 12 -6 imp:n,e,p=256 $FARTHEST RIGHT SOLUTION AREA (5)
7 1 -7.9 -4  3 imp:n,e,p=1 $ TARGET CYLINDER CELL
8 0 -3  5 fill=100 imp:n,e,p=1 $ DEUTERIUM FILL CELL
9 4 -0.0005 -1  6 imp:n,e,p=1 $ AIR IN TANK
10 0 -3 -5 imp:n,e,p=1 $ cylinder face
11 3 -18.95 -7 8 u=100 imp:n,e,p=1 $ DU/NU TARGET
12 0 -8:7 u=100 imp:n,e,p=1 $ surrounding target void
100 0 2 -101 imp:n,e,p=1 $ VOID AROUND DECK
101 0 101 imp:n,e,p=0 $ END OF UNIVERSE

c SURFACE CARDS---------------------------------------------------------------
1 rcc 0 -8 0 0 150 0 75 $ inner cylinder macro
2 rcc 0 -10 0 0 154 0 77 $ outer cylinder macro
3 rcc 0 -10 -25.0 0 32 0 16 $ target source inner cylinder macro
4 rcc 0 -10 -25.0 0 32.5 0 16.5 $ target source outer cylinder macro
5 py -8 $ front fascia plane
6 pz 25 $ Solution fill plane
7 rcc 0 7 -25.0 0 8 0 3 $ target cylinder to be cut with cone
8 trc 0 7 -25.0 0 4 0 3 1e-05 $ cone cutout for cylindrical target
9 py 6.25 $ PLANE FOR CUTTING SOLUTION INTO PARTS
10 py 22.5 $ PLANE 2 FOR CUTTING SOLUTION INTO PARTS
11 py 62.333333333333336 $ PLANE 3 FOR CUTTING SOLUTION INTO PARTS
```

```
12 py 102.16666666666667 $ PLANE 4 FOR CUTTING SOLUTION INTO PARTS
100 c/y 0 -25.0 0.3 $ Constraint Tube for Gaussian (used for CCC cookie cutter cell)
101 rpp -500 500 -500 500 -500 500

c MATERIAL CARDS----------------------------------------------------------------
mode n e p
nps 1e4
phys:p 100 J J 1 J J 0
c prdmp 1e3 1e3 0 20 1e3
prdmp
sdef dir=1 vec 0. 1. 0. x=d1 y=-33 z=d2 erg=40 par=e ccc=100
sp1  -41  0.25479654  0 $ X gaussian spread, 0.6=fy=2.35482a
sp2  -41  0.25479654 -25.0 $ Z gaussian spread, 0.6=fz=2.35482b
f24:n 2 3 4 5 6 T
fm24 (-1 2 -6)
sd24 1 1 1 1 1 1
c fmesh4:n geom=xyz origin=-75 -10 -75
c          imesh=75 iints=160
c          jmesh=140 jints=200
c          kmesh=75 kints=160
c FM4 (-1 2 -6)
c ksrc 0 13.0 -7.5
c kcode 5000 1.0 50 250
c metal (boron 5010 and phos 15031 stripped for physics issue)
m1    14028.80c -0.001150761 16034.80c -2.33827E-06
      24053.80c -0.005492588 26054.80c -0.012563171 26058.80c -0.000645696
      28061.80c -0.000474334 42092.80c -0.001187171 42096.80c -0.001392605
      42100.80c -0.000837433 14029.80c -6.04609E-05 16032.80c -4.74334E-05
      24050.80c -0.002372672 24054.80c -0.001390266 26056.80c -0.204337425
      28058.80c -0.026936829 28062.80c -0.001535239 42094.80c -0.000756262
```

42097.80c -0.0008057 14030.80c -4.14207E-05

16033.80c -3.34038E-07 24052.80c -0.047530943 25055.80c -0.005010571

26057.80c -0.004805805 28060.80c -0.010731975 28064.80c -0.00040619

42095.80c -0.001315108 42098.80c -0.002056672

c heavy water uranyl solution with concetration of 80g/100ml (NU)

C m2 92235.80c 0.007 92238.80c 0.993 01002.80c 24.6237

c m2 92235.80c 0.05 92238.80c 0.95 01002.80c 54.70

C 07014.80c 2 08016.80c 20.3118

C mt2 hwtr.10t

m2 92235.80c 0.007 92238.80c 0.993

01001.80c 99.5891

08016.80c 57.7946

07014.80c 2

mt2 lwtr.10t

MX2:p j j 0 j j

c NU target (18.95 g/cm3)

m3 92235.80c -0.007 92238.80c -0.993

c air (wrong needs adjusting to proper air)

m4 8016.80c 1 7014.80c 2